EXTRUSION AND EXTRUDERS

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INTRODUCTION

Extrusion is the process of forming a raw material into a product of uniform shape and density by forcing it through an orifice or die under controlled conditions. An extruder consists of two distinct parts: a delivery system which transports the material and sometimes imparts a degree of distributive mixing, and a die system which forms the material into the required shape. Extrusion may be broadly classified into molten systems under temperature control or semisolid viscous systems. In molten extrusion, heat is applied to the material in order to control its viscosity to enable it to flow through the die. Semisolid systems are multiphase concentrated dispersions containing a high proportion of solids mixed with a liquid phase. Extrusion is achieved by formulation to control the viscosity of the semisolid mass.

Extrusion is a continuous process that affords a consistent product at high throughput rates. The process has diverse applications in a range of industries utilizing extrusion equipment specifically designed or adapted to form a particular product. A description of the different types of extruders is given here, along with details that illustrate the versatility of extrusion processing.

THEORY AND CHARACTERIZATION OF EXTRUSION

The various types of extruders have the common feature of forcing the extrudate from a wide cross section through the restriction of the die. The force required and the characteristics of the extrudate produced are dependent on the rheological properties of the extrudate, the design of the die, and the rate at which the material is forced through the die. The theoretical approach to understanding the systems, therefore, is generally associated with dividing the process of flow into three sections: 1) entry into the die, 2) flow through the die, and 3) exit from the die.

Extrusion is dependent on the material, and the technique varies with the material studied. With regard to pharmaceuticals, most systems consist of particles dispersed in a fluid and, although consideration will be given to plastics, the main emphasis is on paste extrusions. These differ in the fact that a fluid is present between solid particles. The relative position of solid and liquid can change during the various stages of the extrusion process, and hence produce effects different from those associated with single-phase systems.

If the die is considered as a simple capillary flow, the relationship between the rate of shear (γ) and die wall shear stress (τ_w) can be described by Eq. 1:

$$\tau_w = \gamma P \cdot R/2L \tag{1}$$

where P is the pressure drop across the length of capillary L and radius R. Corrections for entrance effects modify this equation by considering an increase in the length of the capillary to give Eq. 2:

$$\tau_w = \gamma P/2(L/R + n_b) \tag{2}$$

where n_b is the Bagley entrance correction (1). Determination of n_b can be made by measuring the pressure necessary to force extrudate through dies of different length-to-radius (*L*/*R*) ratio. Extrapolation of the graphs to zero pressure values gives the value of n_b as the intercept on the *L*/*R* axis (1).

Han and Charles (2) found experimentally that the exit pressure is actually above atmospheric pressure and proposed modification of Eq. 2 to Eq. 3 corrected for exit pressure losses:

$$\tau_w = (\gamma P - P_e)/2(L/R + n_b) \tag{3}$$

where P_e is the exit pressure. This value is difficult to determine and, because it is considerably lower than the pressure loss upstream and through the die, it is usually neglected. The upstream pressure loss can be considerable and can be determined as the intercept on the pressure axis at zero *L*/*R* ratio (the Bagley equation), giving Eqs. 4 and 5:

$$\tau_w = (P_T - P_0)R/2L \tag{4}$$

$$P_T = P_0 + 2\tau_w(L/R + n_b) \tag{5}$$

The upstream pressure loss includes pressure losses due to kinetic energy, head effects, elastic losses, and turbulence. Harrison (3) found for a series of pharmaceutical systems that the value of P_0 increases with increasing rate of passage through the die.

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Rheological Curves

In addition to determination of the upstream pressure loss P_0 and the end correction n_b, Eq. 5 can be seen to provide a value for the shear stress τ_w in such systems. The slope of the graph P_T versus. L/R has a gradient of $2\tau_w$; that is, the die-wall shear stress. The rate of shear at the die wall—$(dv/dr)_w$—can be derived from the Hagen–Poiseuille's law, as in Eq. 6:

$$(\mathrm{d}v/\mathrm{d}r)_\mathrm{w} = 4Q/R^3 \qquad (6)$$

where Q is the volumetric flow rate and R the radius of the die. This assumes that the flow is Newtonian. If this is not the case, Jastrzebski (4) suggested that a correction should be made for the rate of shear, as in Eq. 7:

$$-(\mathrm{d}v/\mathrm{d}r)_\mathrm{w} = \left(\frac{3n' + 1}{n'}\right)\frac{Q}{\pi R^3} \qquad (7)$$

where n' is the degree of non-Newtonian flow; it is determined from the gradient of the graph of log-shear stress as a function of the log apparent shear rate. Wilkinson (5) has also indicated that these equations assume that: 1) the flow is laminar, 2) there is no slip at the die wall, and 3) the rate of shear depends only on the shear stress at the point of measurement and is independent of time.

Determination of shear rate versus shear stress curves by application of the ram extruder allow characterization of the rheological properties of the extruded material according to the basic type of curve, as expressed by Eqs. 8–11.

Newtonian:

$$\sigma_{\mathrm{w}=\gamma'}\eta \qquad (8)$$

where η is the apparent viscosity and γ' is the rate of shear.

Bingham body:

$$\tau_\mathrm{w} = \sigma_\mathrm{y} + \gamma' U \qquad (9)$$

where σ_y is the stress necessary to be exceeded before Newtonian flow commences, yield value, and U is the plastic viscosity.

Power-law model:

$$\tau_\mathrm{w} = K\gamma^{n'} \qquad (10)$$

where K is the power-law viscosity constant and n' is the degree of non-Newtonian flow. For values of n' less than 1, the material becomes less viscous with increasing shear rate (shear thinning), and for values of n' greater then 1, the viscosity increases with increasing shear rate (shear thickening).

Herschel–Buckley model:

$$\tau_\mathrm{w} = \tau_\mathrm{y} + K\gamma^n \qquad (11)$$

which allows for a system that has a yield value and a shear rate dependent on viscosity.

The application of these types of flow curves requires homogeneous materials that do not change in consistency with extrusion. Harrison et al. (6) found this not to be the case, and suggested that this was due to the presence of plug flow within the extrudate bulk and slip flow at the die wall.

The inability of the standard rheological models to quantitatively describe the process of flow into, through, and out of the die requires an alternative treatment. From a study of ceramic catalyst pastes, Benbow and Ovensten (7) and Benbow (8) assumed that there was broad plug flow at the center of the extrudate, with shearing occurring within a thin liquid layer at the die wall. Assuming that this layer behaves as a Newtonian liquid of thickness x and viscosity η, and that the initial shear stress to induce flow is τ_0, the total die-wall shear stress τ_w at a given extrudate velocity V is given by Eq. 12:

$$\tau_\mathrm{w} = \tau_0 + (\eta/x)V \qquad (12)$$

The values of η and x cannot be determined directly and, therefore, Benbow et al. (9) introduced the term β, the die land viscosity factor, to replace η/x, as in Eq. 13:

$$\tau_\mathrm{w} = \tau_0 = \beta V \qquad (13)$$

Incorporation of this expression into Eq. 4 yields:

$$P_\tau = P_0 + 2(L/R)(\tau_0 + \beta V) \qquad (14)$$

The value of τ_0 can be determined by plotting the extrusion pressure against the extrudate velocity V for extrusion through dies of constant value of L. The extrapolated value of extrusion pressure at $V = 0$ gives the value P_{0v0} at zero velocity, as shown by Eq. 15:

$$P_{\tau v0} = P_{0v0} + 2(L/R)\tau_0 \qquad (15)$$

Thus, a graph of $P_{\tau v0}$ versus L/R provides the value of τ_0 as equal to half the slope. The value of β can be calculated by rearranging Eq. 14 to Eq. 16:

$$\beta = \left(P_{\tau_\mathrm{w}} - P_{0\mathrm{w}}\right) - \left(P_{\tau v0} - P_{0v0}\right)2(L/R)V \qquad (16)$$

where P_{τ_w} is the total extrusion pressure at extrudate velocity V, and P_{0_w} is the upstream pressure loss at extrudate velocity V.

Further characterization of the system was suggested by Benbow (8) and Benbow and Bridgwater (10) in terms of a yield value σ_y associated with the convergence of flow

from the wide cross section of the feed to the narrow cross section of the die. This takes the form of Eq. 17:

$$P_0 = \sigma_y \ln(A_0/A) \tag{17}$$

where A_0 is the initial cross-sectional area and A that of the die. If the original and final cross sections are circular, Eq. 18 holds:

$$P_0 = 2\sigma_y \ln(D_0/D) \tag{18}$$

where D_0 and D are the barrel and die diameters, respectively. For materials that deform plastically and are time independent, the value of σ_y can be calculated from the intercept of the pressure axis divided by twice the natural log reduction ratio (D_0/D) for plots of P against L/R.

By combining this concept with those expressed above, Benbow et al. (9) and Benbow and Bridgwater (10) further modified the Bagley equation to Eq. 19:

$$P_T = 2(\sigma_{y0} + \alpha V) \ln(D_0/D) + 2(L/R)(\tau_0 + \beta V) \tag{19}$$

If the die land velocity factor β varies with the extrusion rate or the liquid layer at the die wall is non-Newtonian, Eq. 19 must be further modified to Eq. 20:

$$P_\tau = 2\sigma_y \ln(D_0/D) + 2(L/R)(\tau_0 + \beta^* V^{l-n}) \tag{20}$$

where β^* is a modified power-law constant and n the degree of non-Newtonian flow. If the flow velocity into the die is also dependent on the velocity of flow, Benbow et al. (9) and Benbow and Bridgwater (10) propose replacement of the yield value δ_y, by two empirical parameters, the initial die entry yield stress σ_{y0} and the die-entry yield-stress velocity factor α. Substituting in Eq. 18 gives Eq. 21:

$$P_0 = 2(\sigma_{y0} + \alpha V) \ln(D_0/D) \tag{21}$$

The fully corrected Bagley equation now becomes Eq. 22:

$$P_T = 2(\sigma_{y0} + \alpha V) \ln(D_0/D) + 2(L/R)(\tau_0 + \beta V) \tag{22}$$

The value of σ_{y0} can be obtained as the intercept from the derived zero-velocity graph of P_{0v0} as a function of L/R. The value of α, for a given system, is obtained from Eq. 23:

$$\alpha = \left(P_{0_w} - P_{v0}\right)/(2 \ln(D_0/D)V) \tag{23}$$

where P_{0_w} and P_{0v0} are obtained as described previously.

If the systems are treated as polymer melts instead of as paste (i.e., homogenous systems with no fluid migration during extrusion), further characterization of the wet masses can be achieved (11). The flow of melts through a capillary rheometer can be considered to show flow streamlines converging and then accelerating, which according to Cogswell (12), is extensional flow. He separated the flow field into shear and tensile deformation and then described their calculation from the following equations:

$$TS = \frac{3}{8}(n+1)P_0 \tag{24}$$

where TS is the tensile stress (i.e., stretching), n is the power law index, and P_0 is the die-entrance press drop,

$$ESR = \frac{4\pi\gamma}{3(n+1)} = \frac{\gamma}{2} \tan\theta \tag{25}$$

where ESR is the tensile stretch rate, τ is the shear stress at the die wall, γ is the shear strain rate, and θ is the half angle of natural convergence.

$$EV \frac{TS}{ESR} \tag{26}$$

where EV is the apparent extensional (elongational) viscosity.

Such an approach depends on the flow fitting the power law model (i.e., Eq. 10), and that flow is not dominated by wall slip.

In addition to elongation flow, material can also exhibit elastic behavior. Two parameters that have been proposed (13, 14) to quantify this property are (1) recoverable shear RS and (2) compliance C. These can be derived from:

$$RS = \frac{P}{4\pi} \tag{27}$$

and

$$C = \frac{P}{4\tau^2} \tag{28}$$

Chohan (14) has used these to study the flow of branched polyethylene melt, and while what is exactly implied by these terms at high stretch rates is not clear, they are undoubtedly related to the elastic behavior of the material. The higher the values of each, the greater will be the elastic nature of the material.

MEASUREMENT OF RHEOLOGICAL PROPERTIES

The application of the theoretical treatment depends on the ability to measure the extrusion force and rate. Most commercial extruders do not allow for these types of measurement. Normal rheological equipment, such as cup-and-bob or cone-and-plate, do not have a suitable

geometry or instrumentation to handle materials of the consistency normally used. A ram extruder is a suitable experimental design.

The ram extruder, designed by Benbow and Ovenston (7), operates on a prefilled system and is used for experimental and small-scale extrusion (Fig. 1). It consists of a stainless steel barrel (2.54 cm internal diameter, approximately 20 cm in length), which acts as the material reservoir. The base is constructed to enable interchangeable dies, with central capillaries of varying dimensions to be bolted on. A rubber ring is inserted between the barrel and die to ensure a watertight connection. The piston, or ram, is a stainless steel rod that fits loosely into the barrel. A fluon ring positioned at its lower end provides a low-friction seal to prevent material escaping above the point where the piston moves down the barrel. The extrusion is a noncontinuous operation; first the material (50–100 g) is packed into the barrel and partially consolidated to a plug by inserting the piston. It is possible to add temperature control to the barrel to extrude materials that are thermosensitive. The barrel-and-die assembly is mounted on a rigid metal C-piece,

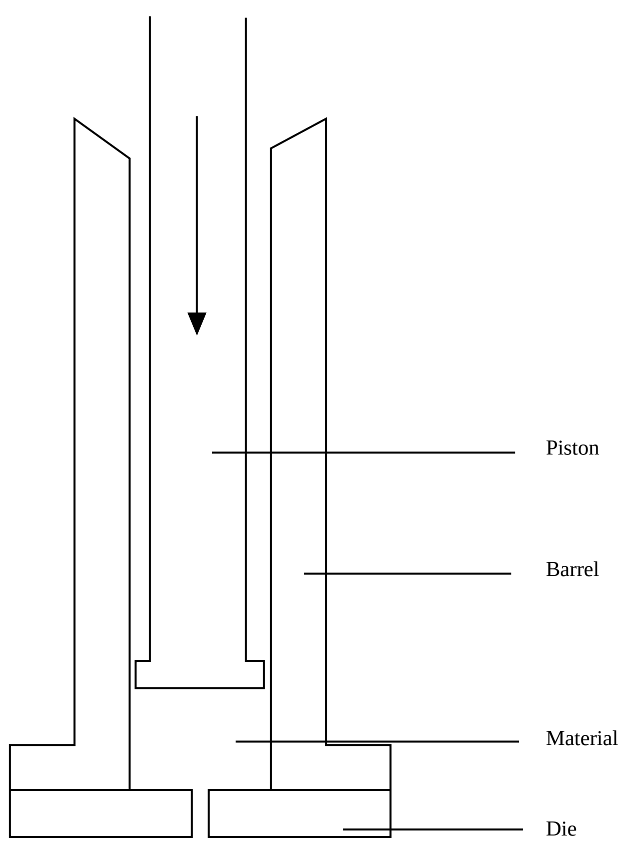

Fig. 1 Diagram of a ram extruder.

and a load is applied to the piston sufficient to extrude the material through the die. The ram extruder can be used in conjunction with an instrumented press. The piston is attached to the cross-head that may be driven down at various constant rates, and its displacement monitored by an attached displacement transducer. Output from this and the load cell is fed into an $x-y$ chart recorder or computer. This arrangement enables the force acting on the material during extrusion to be recorded as a function of the displacement of the piston, and a force–displacement profile is produced.

A typical extrusion mixture produces three distinct regions, as shown in Fig. 2. In the compression stage, the piston descends into the barrel and consolidates the material into a plug prior to flow. This results in a large change in displacement accompanied by a small change in load. Eventually, the material is compressed to its minimum volume and maximum density. At this point, the pressure builds up while the material density is maintained. This is shown in the profile by a large increase in load accompanied by a minimal change in displacement. At the end of the compression stage, the pressure applied to the mass increases until it is high enough for the material to yield and commence flow. This is followed by a period of steady-state flow in which the force required to maintain the extrusion remains constant as the displacement increases. Forced flow occurs when steady-state flow can no longer be maintained. It leads to a gradual rise in extrusion force with displacement. This occurs often toward the end of the extrusion and is caused by the close proximity of the ram tip to the die face.

The force–displacement profile is altered by varying one of the extrusion parameters, such as the die diameter, *L*/*R*, or extrusion rate. For a given mixture, the relationship between the steady-state extrusion force, the die *L*/*R* at constant die diameter, and the extrusion rate can be represented graphically, as shown in Fig. 3. This is known as the Bagley plot (1). Used in this way, the extruder operates on a principle similar to that of a capillary rheometer, and expressions derived from capillary rheometry (described previously) may be used to characterize the properties of the wet powder mass. After conversion of the steady-state force values to pressure values, the slope of the relationship between the pressure and *L*/*R* is numerically equivalent to twice the value of the mean die-wall shear stress (according to Eq. 4). Plotting these values against the corresponding apparent die-wall shear rates (derived from Eq. 6) results in a flow curve that is unique for a particular wet-mass formulation (Fig. 4). The materials exhibit non-Newtonian flow and shear-thinning properties.

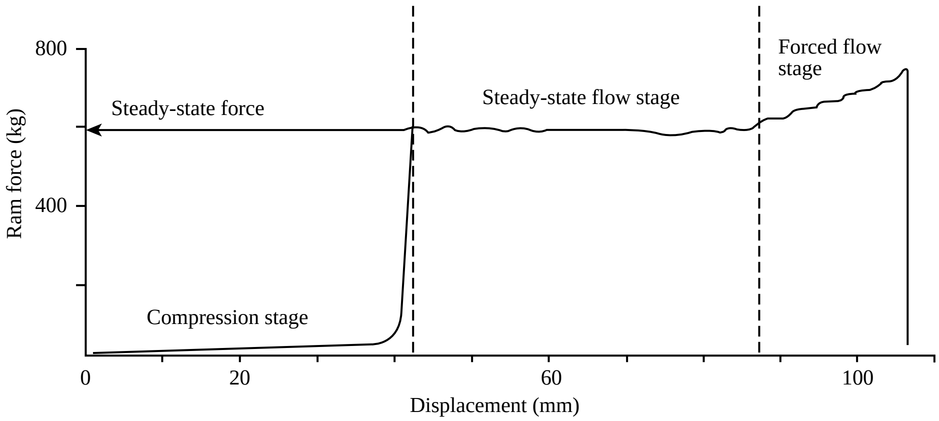

Fig. 2 Force–displacement profile for microcrystalline cellulose–lactose–water mixture.

PRACTICAL CHARACTERIZATION OF EXTRUSION SYSTEMS

The expression of the extrusion properties of pharmaceutical systems by numerical values could aid formulation. To be able to apply the theoretical approaches described previously, it is important to ensure that the restrictions of the systems are considered. One major problem with paste systems is that when subjected to pressure, there is phase separation resulting in variations in the composition of the mass as it is being extruded. This can be detected by collecting the extrudate and measuring its water content (15, 16). Alternatively, magnetic resonance imaging has been used to quantify the water distribution within the barrel (17) and within the extrudate (18).

The extent to which die-wall slip is involved can be assessed by using dies of different lengths and diameters. An important characteristic that can be observed in the extrudate is its quality in terms of surface structure. Harrison et al. (19) have shown how this can vary from a smooth, regular surface via a rough, "shark-skinned" extrudate. There is obvious need to prevent this

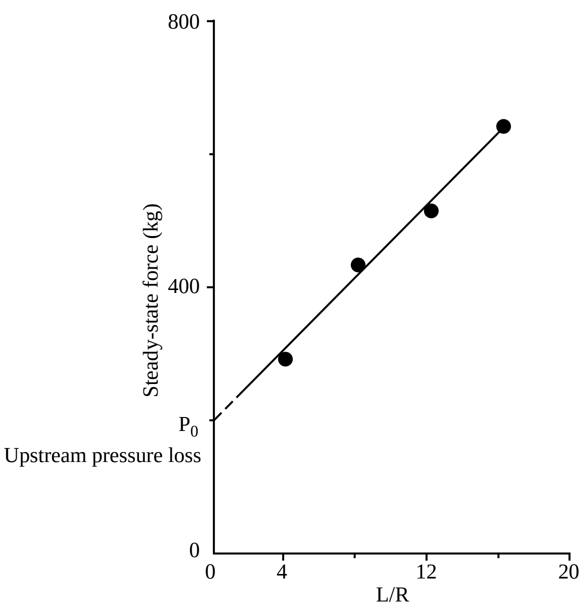

Fig. 3 Steady-state extrusion force as a function of the length-to-radius ratio of the die for microcrystalline cellulose–lactose–water (5:5:6) at constant die diameter (1.5 mm) and extrusion rate (20 cm/min).

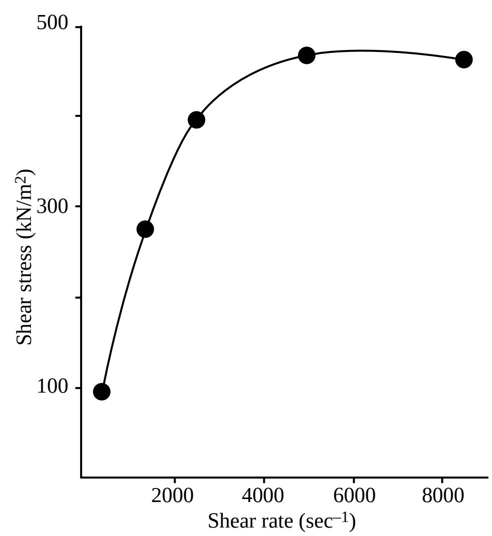

Fig. 4 Typical shear stress-shear rate flow curve for an extrusion mixture containing 50% microcrystalline cellulose extruded through a 1.5-mm diameter die.

phenomenon if extrudate of the correct quality is to be produced. The occurrence of the surface defects is associated with both the composition of the material and the operating conditions (e.g., die length and diameter and the rate of extrusion) (Fig. 5). Raines et al. (20) were able to relate the quality of the surface of the extrudate to the value of the yield stress at zero velocity δ_{y0}, in that those systems with a high value were smooth and regular while those with low values were shark-skinned.

Of the systems studied, most pastes show non-Newtonian behavior. This has important consequences for extruder design and operating conditions as material that are shear-rate dependent require careful handling. To date, most reported rheological investigations indicate that paste systems are shear thinning (i.e., their viscosity decreases with an increase in shear rate) (Fig. 4). Their extent of property can be quantified by obtaining the value of n', the slope of the log shear-rate/log shear-stress graph. There is also some evidence that paste systems show plug flow (i.e., the central core of the extrudate moves at a constant velocity), while there is a thin layer of moisture at the die wall where shear takes place (21).

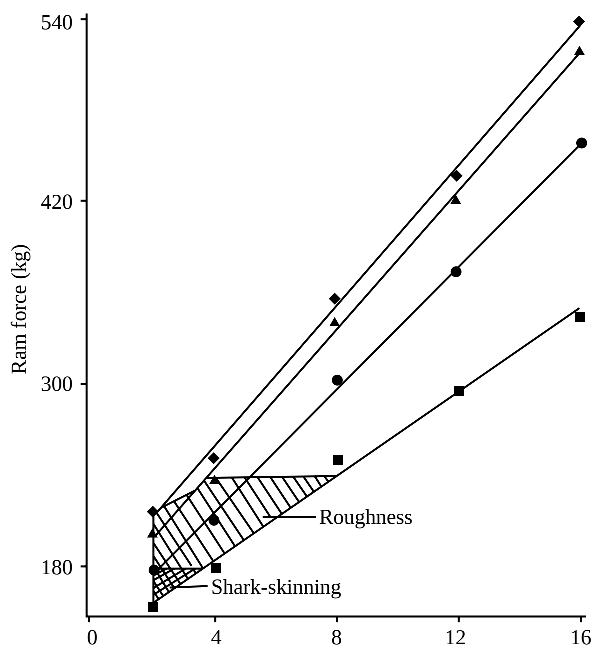

Fig. 5 A graph of ram force as a function of length-to-radius ratio, depicting conditions under which surface defects occur when extruding microcrystalline cellulose–lactose–water (5:5:6). Die diameter = 1.5 mm; ram speed cm/min: ■, 5; ●, 10; ▲, 20; ◆, 40.

FORMULATION

Extrusion mixtures are formulated to produce a cohesive plastic mass that remains homogeneous during extrusion. The mass must possess inherent fluidity, permitting flow during the process and self-lubricating properties as it passes through the die. The resultant extrudate must remain nonadhesive to itself and retain a degree of rigidity so that the shape imposed by the die is retained. Precise formulation requirements depend upon subsequent processing. Extrudate that is simply to be cut to short lengths to form cylindrical granules that are dried in a fluid-bed drier can be less rigid than extrudate intended for complex processing such as spheronization, where the extrudate undergoes a series of subtle shape changes.

The requirements for spheronization of the cylindrical extrudate are as follows:

1. The extrudate must possess sufficient mechanical strength when wet, yet it must be brittle enough to be broken down to short lengths in the spheronizer, but not to be so friable that it disintegrates completely. To achieve a narrow size distribution of spheres, the extrudate is ideally reduced to cylindrical rods of uniform length equal to approximately one and a half times their diameter (22).
2. The extrudate must be sufficiently plastic to enable the cylindrical rods to be rolled into spheres by the action of the friction plate in the spheronizer.
3. The extrudate must be nonadhesive to itself in order that each spherical granule remains discrete throughout the process.

A typical extrusion mixture might contain the following ingredients:

Drug	50–90%
Extrusion aid	
Microcrystalline cellulose, bentonite	5–90%
Binder	
Polyvinylpyrrolidone (PVP)	
Sodium carboxymethylcellulose (SCMC)	
Hydroxypropyl methylcellulose (HPMC)	
Fluid	
Water or solvent	

Extrusion offers the advantage of incorporating a relatively high proportion of active ingredient, up to 90%, in the final product. However, the physicochemical properties of the drug determine to a large extent the maximum quantity that can be included in a particular formulation. An extrusion aid is essential; microcrystalline cellulose is commonly used (23). The function of

microcrystalline cellulose is two-fold: it controls the movement of water through the wet powder mass during extrusion, and modifies the rheological properties of the other ingredients in the mixture, conferring a degree of plasticity which allows it to be readily extruded. This interaction with the liquid phase is both a physical and chemical phenomenon. The microscopic structure of microcrystalline cellulose is a random aggregation of filamentous microcrystals that create a high internal porosity and a large surface area, approximately 130–270 m^2/g (24). This provides highly absorbent and moisture-retaining characteristics that are often unaffected by the extrusion process. This could be the essential quality that makes microcrystalline cellulose a unique material for extrusion. Bentonite and kaolin also have been used. Inclusion of 5–10% can significantly improve the extrusion properties of mixtures containing high proportions of drug. Recent work (25) has shown that it is possible to reduce the quantity of microcrystalline cellulose by adding glyceryl monostearate.

Additional ingredients may or may not be necessary. A binder increases plasticity and reduces extrudate friability, particularly when the content of microcrystalline cellulose is low. Natural or synthetic polymers, such as gelatin, PVP, or SCMC, may be incorporated into the mixture as a solid during dry mixing or in solution in the liquid phase. Commercial preparations of microcrystalline cellulose that are already combined with polymers are available. Examples include Avicel RC and Avicel CL grades of microcrystalline cellulose combined with SCMC (FMC Corporation). Variations in the type of microcrystalline cellulose significantly change the rheological properties of the mixture, and therefore, the extrusion characteristics (20). The differences between shear stress–shear rate flow curves of mixtures of microcrystalline cellulose–lactose–water (5:5:6) containing different particle sizes of microcrystalline cellulose and different quantities of SCMC are distinct but different. Inclusion of a polymer in the wet mass produces marked rheological differences. This has implications in the choice of formulations, since the extrudates formed from these various microcrystalline cellulose mixtures behave differently during subsequent processing, such as cutting, spheronization, and drying.

The mixture of dry ingredients is blended with water or a solvent such as ethanol (26) to form a dense cohesive mass suitable for extrusion. The liquid content of the wet powder mass and its distribution are highly critical and should be controlled so that they produce an extrudate that posses the ideal characteristics. In general, these wet mixes have a much higher moisture content, typically 20–30 wt%, than is required for conventional (tablet) granulations, the aim being to produce as dense a material as possible for passing through the extruder. Fluffy and incompletely wetted masses feed poorly and cause problems by creating excessive pressure and friction within the equipment. On spheronizing, they tend to produce large quantities of fines, and the "dry" extrudate is insufficiently plastic, forming dumbbell-shaped or ovoid pellets which never round off into spheres. On the other hand, if the mixture is too wet, it produces an extrudate that adheres to the spheronizer plate and to itself. This product tends to aggregate uncontrollably or at best produce spheres of wide-size distribution as the material is transferred from pellet to pellet via the plate motion.

The possible processability of different drugs by this approach has not yet been fully established. It is not possible to relate the pK_a, and freezing point depression, or to relate the ability to produce uniform pellets from a spheronization grade of microcrystalline cellulose (27). However, a relationship between the water solubility and the water level required by a formulation for equal parts of a series of model drugs and microcrystalline cellulose has been established (28).

INDUSTRIAL APPLICATIONS

Plastics

Extrusion technology is extensively applied in the plastics and rubber industries where it is one of the most important fabrication processes. Examples of products made from extruded polymers include pipes, hoses, insulated wires and cables, plastic and rubber sheeting, and polystyrene tiles. The most common extruder employed is the single-screw type (Fig. 6) with either cold or hot feed, which requires the polymer to be heated prior to processing. The extruder consists of a rotating screw inside a stationary cylindrical barrel. The barrel is often manufactured in sections that are bolted or clamped together. Usually, the inner surface of the barrel is grooved to reduce slippage and increase pumping capability. An end-plate die, connected to the end of the barrel, determines the configuration of the extruded product.

The extruder is conventionally divided into three sections: feed zone, transition zone, and metering zone. Resin granules are fed from a hopper directly into the feed section, which has deeper flights or flights of greater pitch. This geometry enables the feed material to fall easily into the screw for conveying along the barrel. The pellets are transported as a solid plug to the transition zone where they are mixed, compressed, melted, and

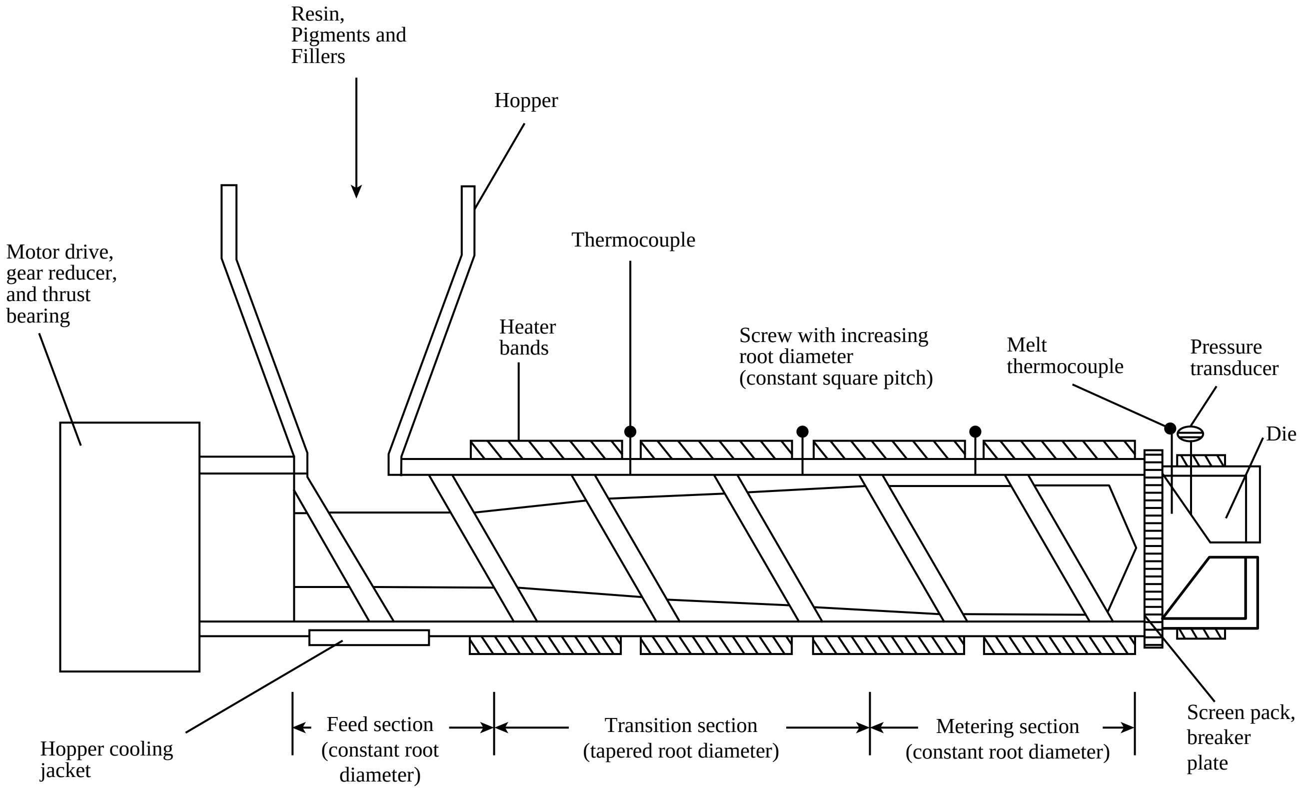

Fig. 6 Component parts of a single-screw extruder.

plasticized. Compression is developed by decreasing the thread pitch but maintaining a constant flight depth or by decreasing flight depth while maintaining a constant thread pitch (29). Both methods result in increased pressure as the material moves along the barrel. Most of the heat required to melt the material is supplied by the heat generated by friction as the resin granules are sheared between the rotating screw and the wall of the barrel. Additional heat may be supplied by electric heaters mounted on the barrel. The melt moves by circulation in a helical path by means of transverse flow, drag flow, pressure flow, and leakage: the latter two mechanisms reverse the flow of material along the barrel. The material reaches the metering zone in the form of a homogeneous plastic melt suitable for extrusion. For an extrudate of uniform thickness, flow must be consistent and without stagnant zones right up to the die entrance. The function of the metering zone is to reduce pulsating flow and ensure a uniform delivery rate through the die cavity. Some applications require a strainer plate fitted between the extruder and die plate to remove solid impurities or lumps of incompletely melted resin.

Polymers with a wide range of viscoelastic and melt viscosities cannot be processed with a single screw. Most commercial extruders are, therefore, modular in design, providing a choice of screws or interchangeable sections that alter the configuration of the feed, transition, and metering zones. This makes it possible to modify the process to meet particular requirements, for example, from a standard to a high shear or high output extrusion. Modified screw designs allow the extruder to perform a mixing role in addition to extrusion, so that the material can be colored and blended. The various screw and die designs available and practical considerations of thermoplastic extrusion are reviewed by Whelan and Dunning (30). Extrusion processing requires close monitoring of the various parameters that affect polymer extrusion: viscosity, variation of viscosity with shear rate and temperature, elasticity, extensional flow, and slippage of the material over hot metal surfaces. Equations used to describe flow are included in the section on the "Theory and Characterization of Extrusion" presented earlier. Recent advances in the design and operation of extruders allow in-process monitoring and control of parameters, such as the temperature in the extruder, head, and die; pressures in extruder and die; wall thickness and other dimensions; "haul-off" speed and extrusion speed; and power consumption.

The process described above is known as profile or line extrusion in which the shape of the extrudate is determined by the die. The extruded profile proceeds horizontally to the cutoff equipment, which controls its length. It is then cooled to a solid state, usually by spraying with or immersion in water, and passed through a haul-off unit. Finally, it is cut to the required length or coiled. The downstream auxiliaries (e.g., such as haul-off equipment for handling the extrudate stream, collection machinery for winding or coiling continuous lengths of tubing or profiles, cropping and cooling equipment, and systems for monitoring the diameter and wall thicknesses of pipes on-line) are as important as the extruder itself (30, 31). Tubes and pipes and other solid cross-sections are mainly produced by profile extrusion. Profiles may be further processed, for example, as in film extrusion, blow molding, or injection molding (32).

Film extrusion

The polymer melt is extruded through a long slit die onto highly polished cooled rolls that form and wind the finished sheet. This is known as cast film. Plastic packaging film is also formed by blow extrusion, where tubular film is produced by extruding the melt, usually vertically, through an annular-shaped slit die. The extruded tube is inflated by air to form a large cylinder. The bubble is cooled externally by an airstream directed onto its surface and is collapsed on passing between a pair of rollers before being wound up. Film made by the casting process generally has better optical properties than blown film, but is less strong mechanically. Cast films usually require edge trimming at additional cost.

Blow molding

The plastic is heated to a melted or viscous state and a section of molten polymer tubing (parison) is extruded usually downward from the die head into an open mold. The mold is closed around the parison, sealing it at one end. Compressed air is blown into the open end of the tube, expanding the viscous plastic to the walls of the cavity, thus forming the desired shape of the container. The material cools in the cavity and solidifies. The mold is opened and the molding is removed. This technique is used for the manufacture of bottles, toys, and large containers.

Injection molding

The molten plastic is extruded into a cavity mold at high pressure. The material cools in the cavity and solidifies. The mold is then opened and the article is removed. Very intricate configurations can be obtained by this technique (e.g., to provide intricate and strong components for the electronic, telecommunications, and clock-making industries).

Plastics that are commonly processed by extrusion include acrylics (polymethacrylates, polyacrylates) and copolymers of acrylonitrile; cellulosics (cellulose acetate, propionate, and acetate butyrate); polyethylene (low and high density); polypropylene; polystyrene; vinyl plastics; polycarbonates; and nylons. The material properties and extrusion properties have been reviewed by Whelan and Dunning (30). Additives that may be included to modify or enhance properties (33) include lubricants and antislip agents to assist processing during extrusion; plasticizers to achieve softness and flexibility; stabilizers and antioxidants to retard or prevent degradation; and dyes and pigments.

Food

In principle, any food that can be formed into a paste can be processed by an extruder. Food extrusion has been utilized since the 1930s for pasta production. Modern equipment and processing techniques allow the manu-facture of complex products in a variety of shapes and sizes. Raw materials such as cereals, oil seed, and protein, along with carbohydrates and water mixtures, can be converted into products such as meat substitutes, pet foods, and snack meals. A widely used and versatile technique combines cooking and extrusion in a so-called extrusion cooker (34). It has the potential to manufacture a range of novelty or specialty products, such as breakfast cereals (expanded and shaped cereals), shaped and filled snacks, protein-fortified and precooked pasta products, and precooked meat pieces for convenience foods. The process is highly economical, and provides mixing, high temperature–short duration cooking, texturizing, and shaping of the food in one step. The equipment closely resembles the screw extruders used in the processing of thermoplastics. The screw is designed to create varying zones along the barrel, allowing the food substance to be processed in stages. The solid and liquid starting materials are fed from a hopper to the feed zone of the extruder and conveyed to the transition zone. Here the materials may be compressed, mixed, sheared, and heated to form a viscous plastic dough. In the metering zone, the plastic mass is subjected to further heating and shearing before being pumped into the die to form the shaped product. The pressure drop on leaving the die causes superheated water to flash off the molten material. If the dough contains starch, gelatinization will result in an expanded porous product with a crunchy texture (35). Finally, the product may be cut, shaped by passing through rollers, dried, and packed.

The viscosity of the dough may vary more than an order of magnitude during the extrusion cooking process as a result of changes in shear rate, temperature, moisture content, and induced physicochemical changes such as protein denaturation, polysaccharide gel formation, and reorientation of molecules (36). For this reason, success in food extrusion requires accurate monitoring and control of feed rate, screw speed, temperature, and moisture to produce and control desired product characteristics. Knowledge of the viscous rheology of the food mixture in the metering section immediately prior to extrusion is of particular importance. However, this is not easily predicted since, unlike the case of homogeneous or simple mixtures of polymers where the major change is melting, food doughs are of such complexity that the exact chemical composition and structure cannot readily be determined. Efforts have been made to develop semiempirical models derived from plastics extrusion to describe the apparent viscosity of cooking doughs, which may be useful in evaluating food formulations (37, 38). Remsen and Clarke (36) used an Instron capillary viscometer and amylograph to describe the relationship between the viscosity of a typical soy flour dough and the applied shear rate, temperature, and time–temperature history. Fletcher et al. (39) investigated the viscous dough rheology of maize mixtures as a function of the extrusion variables (pressure, shear, and temperature). They used an instrumented single-screw extruder fitted with slit dies, and related the results to the product properties. The advantage with this method is that the food material receives a deformation history corresponding to the extrusion cooking process, which is otherwise difficult to replicate in a laboratory rheometer.

Animal Feed Production

In the animal feed industry, extrusion is applied as a means of producing pelletized feeds, commonly in the form of short cylindrical rods of 4–8 mm in diameter. Pellets are a convenient means of precisely controlling the animal's diet. They offer several advantages (40). The quantity of feed the animal receives is better controlled by pellets than a loose-mix feed, and a complex diet of controlled composition is easily produced. The pellet feed can contain as many as 30 single ingredients mixed in the correct proportions. The animal is obliged to chew pellet feed with improved palatability and therefore, digestion. During extrusion, the feed mixture is compressed, resulting in a densified product that requires less storage space.

Pellets are prepared from a mixture of raw materials of varying chemical composition (starch, oil, fiber, and moisture) and physical characteristics (particle size, bulk density, and moisture-retention properties). The composition of a typical poultry feed is often complex (41). The raw material properties determine the quality of pellet formation. Equipment performance and pellet quality (friability, size uniformity) can be improved by a small amount of extrusion or pelleting aid (binders, lubricants) (42). Additives commonly used in the feed industry include molasses with binding properties when activated by steam; fatty acid lubricants to reduce product–metal friction when extruding or pressing through long dies; lignosulfates (organic materials derived from lignin in trees that improve pellet quality and throughput rates); and mineral binders, such as ball clay and bentonite, or cellulose binders, such as sodium carboxymethylcellulose. It should be noted that in small quantities cellulose binders can improve the pelleting process and reduce pellet friability.

The pelleting process (40) consists of blending and conditioning the feed mixture immediately prior to pressing, pressing itself, cutting of the pellets, and cooling. The complexity of the feed mixture, composed of a number of ingredients of different particle sizes and densities in varying proportions, requires thorough blending to ensure homogeneity. The product is conditioned by adding moisture, typically up to 15%, and heating to a controlled temperature in order to gelatinize the starch or convert it to simple sugars. This reaction causes the starch to act as a binder and converts the meal into a physical state suitable for pressing. The most efficient means of conditioning and heating is by steam. Optimal conditioning parameters, moisture content of the material, temperature, and duration of heating depend on the composition of the mixture (42). For example, high starch–low fiber meals require temperatures of 80–85°C, whereas feeds that contain heat-sensitive ingredients, such as milk and sugar, have a temperature limit of 55°C (39).

Extrusion Pressing

According to Sebestyen (40), pellet mills may be classified into disk-die presses or ring-die pellet mills. In the former, the die consists of a circular plate resting in the horizontal plane into which holes are drilled in a regular pattern. A set of rollers move around the upper surface of the disk, sweeping the meal in their path through the holes and compressing it to form pellets or cubes. Rotating adjustable knives located beneath the disk cut the extrudate to an appropriate length. In another design, the plate revolves while the rollers and knives remain fixed.

Ring-type pellet mills have a radially arranged die resting in the horizontal plane with rollers rotating and revolving along the inner surface. The rolls are offset from the die face, leaving a slight clearance that allows buildup of a thin product layer, optimizing throughput efficiency. The peripheral velocity of the rollers depends upon the die diameter; that is, higher speeds are required for smaller-diameter holes and lower speeds for larger-diameter holes.

Cooling

On leaving the extruder, the warm pellets are pliable and prone to abrasion and deformation. Therefore, a final processing stage is required to harden the pellets (40). Cooling equipment placed directly beneath the mill employs ambient or chilled air to reduce the temperature and remove excess moisture from the final product.

PHARMACEUTICAL INDUSTRY

Extrusion processes are applied within the pharmaceutical industry to produce a variety of dosage forms such as suppositories, implants, and granulations.

The large-scale manufacture of suppositories and pessaries uses either the fusion method where the drug is dispersed in a molten base and the mixture poured into molds to solidify, or the cold compression method (43, 44). In the latter process, the medicament and cold-grated base, usually theobroma oil or witepsol base, are intimately mixed and placed in a cylinder. The mass is extruded by means of a piston through small holes that connect with the mold. The cavities are filled by pressure with the mass which is prevented from escaping by movable end plates. The plates are removed and the suppositories ejected by further extrusion. The extrusion equipment is chilled to prevent melting of the components due to the heat generated by the friction of compression.

The most important application of extrusion in the pharmaceutical industry is in the preparation of granules or pellets of uniform size, shape, and density that contain one or more drugs. The process involves a preliminary stage in which dry powders, drug, and excipients are mixed by conventional blenders, followed by addition of a liquid phase and further mixing to ensure homogeneous distribution (Fig. 7). The wet powder mass is extruded through cylindrical dies or perforated screens with circular holes, typically 0.5–2.0 mm in diameter, to form cylindrical extrudates. These may be further processed, for example, by cutting and drying to yield granules, or by spheronization (24) to yield spherical granules followed by drying. The spheroids are usually coated with a polymer to control the rate of drug release and filled into hard gelatin capsules to yield a multiple-unit dosage form.

Extruders Used for Pharmaceuticals

Commercial extruders may be classified according to the die design and the feed mechanism that transports the material to the die region.

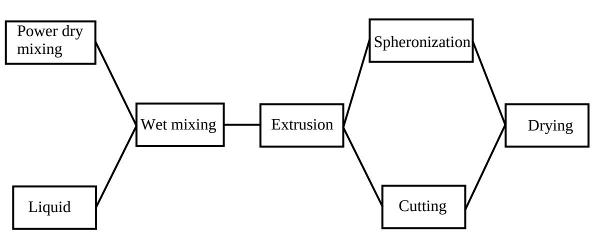

Fig. 7 Schematic of extrusion processing in the pharmaceutical industry.

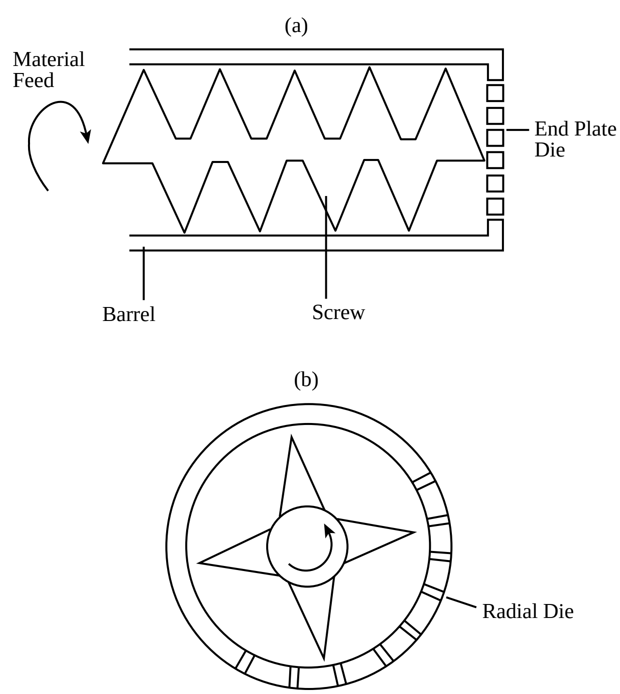

Fig. 8 Screw extruder with (a) end-plate die and (b) radial-screen die.

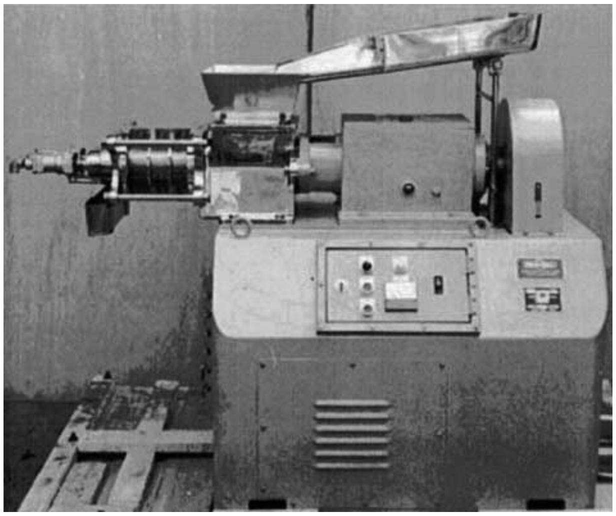

Fig. 9 Twin-screw extruder with radial-screen die. (Manufactured by Fuji Pauldal Company, Japan.)

Screen extruders

Screen extruders utilize a screw-feed mechanism consisting of single or twin helical screws rotating in a barrel to convey the damp mass from a feed hopper to the die zone. The die consists of a thin steel plate perforated with numerous holes, which is positioned radially or axially to the screw feed (Fig. 8). The advantages of this arrangement are high continuous throughput rates, from 5 kg/h of wet mass for a laboratory-scale single-screw extruder, up to 800 kg/h for a larger twin-screw design. The screens are easily cleaned and interchanged; they have holes of varying diameter beginning at 0.5 mm and are available commercially. The disadvantage of this type of equipment, however, is that the screw mechanism can exert a high pressure on the material, generating excessive friction and heat as the wet mass passes between the screw and barrel. This is particularly the case with axially orientated dies. These extruders tend to have a high dead volume that contains stagnant material between the feed screws and the screen. Consideration should be given to this if the wet powder mass contains ingredients that are unstable when wetted with water. The low *L*/*R* of the die holes can also result in low compaction in the extrudate and distortion of the surface finish, known as shark-skinning. This problem can sometimes be overcome by varying the throughput rate, which will be discussed later.

The twin-screw design and radial-die screen assembly of an extruder manufactured by the Fuji Paudal Company of Japan is shown in Fig. 9. Water can be circulated

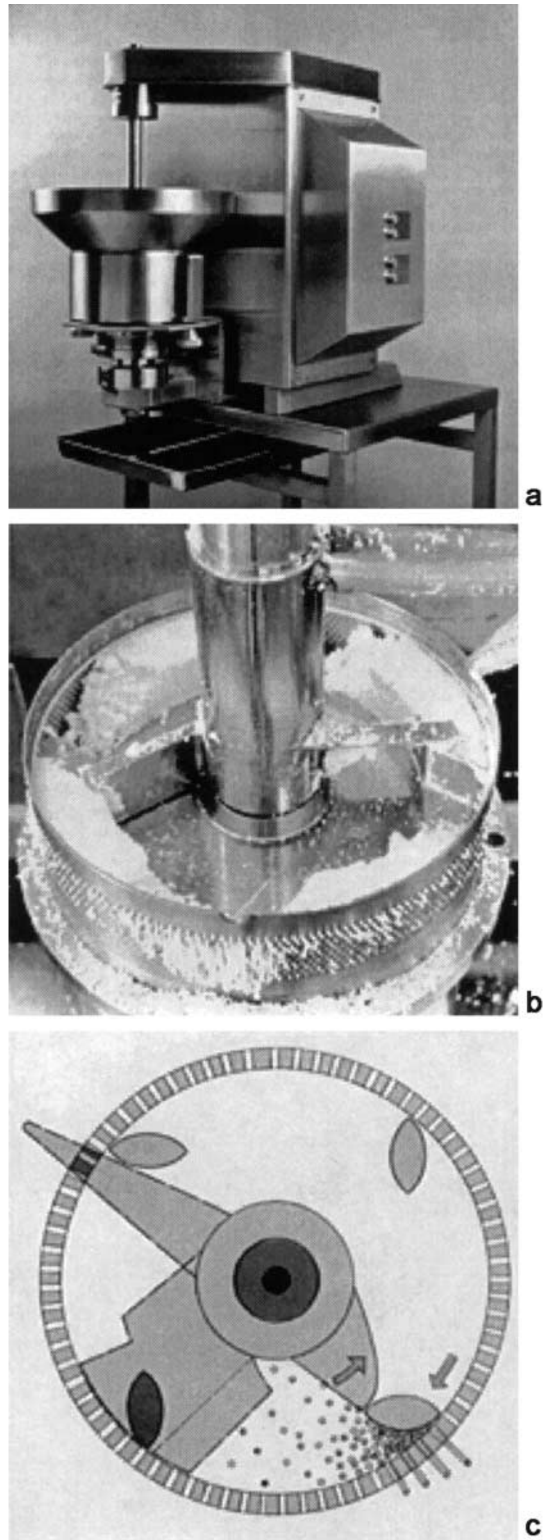

Fig. 10 The NICA system radial-screen extruder. (a) Assembled unit. (b) Dismantled to show extrusion mechanism. (c) Cross section indicating working principle.

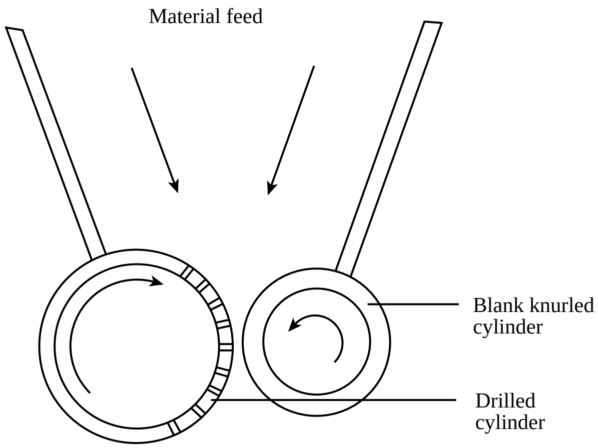

Fig. 11 The rotary-cylinder-type extruder.

through the hollow extrusion rotors to maintain a constant temperature in the extrusion zone. This is a useful facility when processing heat-sensitive materials and for controlling temperature, extrudate moisture levels, and viscosity. Interchangeable screens are available with die holes ranging from 0.5 to 1.5 mm in diameter, allowing the production of a wide range of extrudates. Explosion-proof motors, with fixed or variable speed drive, are fitted for safe processing of wet masses granulated with inflammable solvents. All components in contact with process materials are constructed of high-grade stainless steel. An additional feature is the option of fitting an axial die plate in cases where a denser extrudate is required.

A screen extruder that operates with a novel mechanism is the Nica System Extruder (Fig. 10), manufactured in Sweden. It consists of a radial screen encircling an extrusion rotor and a rotating disk fitted with angled impeller blades or baffles. Above this is a counterrotating central feed blade. Speeds of both the extrusion rotor and the feed blade are variable. Material, such as gravity fed from a hopper, is swept into the blades and pressed through the holes in the screen, According to the manufacturer, there are several advantages to this equipment. First, pressure is exerted on only a small quantity of mass and only at the point of extrusion; that is, just between the baffles and the screen. Second, temperature increase is minimal, and a moisture gradient between the wet mass and extrudate is avoided. Because of this, cooling facilities are not necessary. The dead volume, located in front of each baffle, is limited and may be as low as 15 g per baffle. A small extruder with output up to 4 kg/min is available for development and small-scale production. For larger production, an extruder with output of up to 12 kg/min is available.

Rotary-cylinder extruder

The working principle of this machine is based on two counterrotating cylinders (Fig. 11). The granulating cylinder is perforated and acts as the die. The diameter and the *L*/*R* of the holes can be varied. The holes are spaced further apart and are drilled rather than of punched sheet construction as in the screen-type extruders. The other cylinder is solid and acts as a pressure cylinder. Material is gravity fed from a hopper to the die region between the cylinders and adheres to the knurled surface of the solid cylinder, building up a thin layer that is pressed through the die cylinder. Although the extrusion is a continuous process, actual material flow though each hole is intermittent due to the rotation of the die. Pressure is built up in the perforations, which compacts the wet mass and forces the extrudate to the interior of the cylinder. This pressure is dependent upon the diameter and the length of the perforations. Hence, with die holes of high *L*/*R*, this system can achieve good densification of the wet powder mass. This is important in giving the most granules mechanical strength and stability for further processing. Another advantage is the lack of a dead zone, which is limited to the thin layer of material adhering to the pressure cylinder. Since the cylinders only apply pressure to a small quantity of material in the feed zone, there is little tendency for creating moisture gradients. However, cleaning of the granulating cylinder can be troublesome. The material remaining in the die holes can be difficult to dislodge, particularly when the *L*/*R* is high. Furthermore, the granulating cylinders are expensive because of the high costs of drilling stainless steel.

A cylinder extruder manufactured by Alexanderwerk (Germany) is shown in Fig. 12. With this equipment, the feed stock material can be metered to the working area. On the smallest machines this is accomplished by a rotary-table feed hopper. On larger machines, the feed rate is controlled by screw feeders sited through the feed hopper. The throughput rate depends on the diameter and *L*/*R* of the die holes, as well as on the feed rate. Laboratory-scale extruders with a throughput range of 30 to 50 kg/h use granulation cylinders 70 mm in diameter. Production-scale equipment with a larger granulating cylinder (186 mm diameter) can achieve an output of 100–105 kg/h. Interchangeable cylinders with die holes of 1.0–5.0 mm are available. A multiple-unit assembly consisting of three parallel

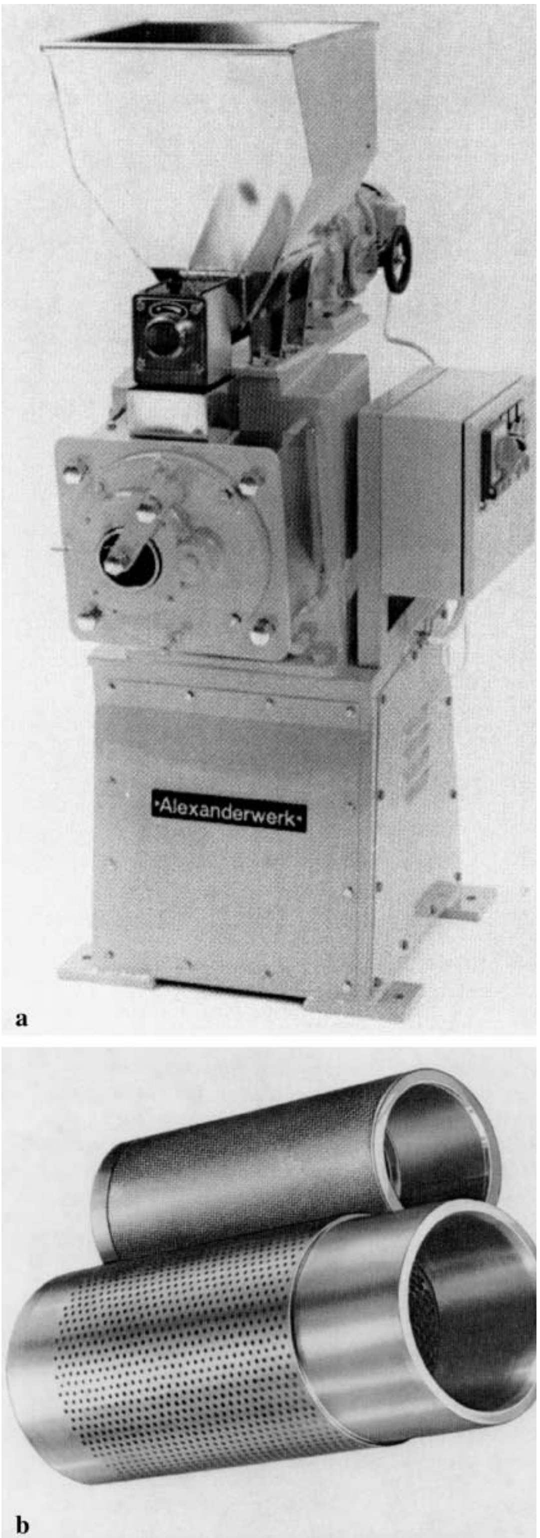

Fig. 12 The cylinder extruder, manufactured by Alexanderwerk, Germany. (a) Laboratory-scale extruder. (b) Die cylinder and pressure cylinder.

extrusion heads can achieve even higher throughput rates of up to 3000 kg/h.

When scaling up production, the die cylinder dimensions cannot be increased proportionately, since the wall thickness and therefore, die *L/R* become too high. This results in excessive extrusion forces imparted on the material. This is overcome by using special counterbored die cylinders with reduced depth perforations to provide optimal extrusion conditions at scale-up. The temperature increase in the extrudate is minimized by circulating cool water through the pressure cylinders. A scraper blade attachment inside the perforated cylinder cuts off the extrudate to shorter lengths, making it more manageable.

Rotary-gear extruder

The rotary-gear extruder operates on a similar concept to that of the cylinder extruder. It consists of two hollow counterrotating gear cylinders with counterbored dies, described by the manufacturer (Bepex, Berwind Corporation) as nozzles, that are drilled into the cylinders between the teeth (Fig. 13). The material, gravity fed from a hopper, is drawn in by the toothed cylinders and pushed through the nozzles into the center of the cylinders, where scrapers cut off the extrudate. The product is compacted as it passes through the nozzles, and thereby forms a dense extrudate. The density depends on the nozzle *L/D* (the ratio of nozzle length to nozzle diameter). Higher throughput rates can be attained with this type of extruder, since output is achieved through both rotating-gear wheels. The equipment and the gear-toothed cylinders are shown in Fig. 13b.

Interchangeable gear cylinders are available with variable nozzle *L/D* ratios by counterboring from the inside of the rollers to reduce the die length or by using replaceable nozzle inserts to increase the die length. The diameter of the holes can be varied from 1.0 to 10.0 mm to produce a range of pellet sizes. The throughput rate can be controlled by varying the cylinders' rotation speed and the corresponding feed rate. Throughput capacity ranges from 20 kg/h for the small-scale laboratory extruders to approximately 1000 kg/h for production equipment. For large equipment or materials with poor flow characteristics, agitators can be installed in the hopper to prevent bridging; special hoppers with conical feed screws fitted with additional wipers are used for highly viscous products. The machine can be furnished with cooling equipment, circulating water through the compaction gears for processing materials that need temperature control. An alternative pharmaceutical gear extruder, similar in design to the above, has recently been marketed by G.B. Caleva Ltd., United Kingdom.

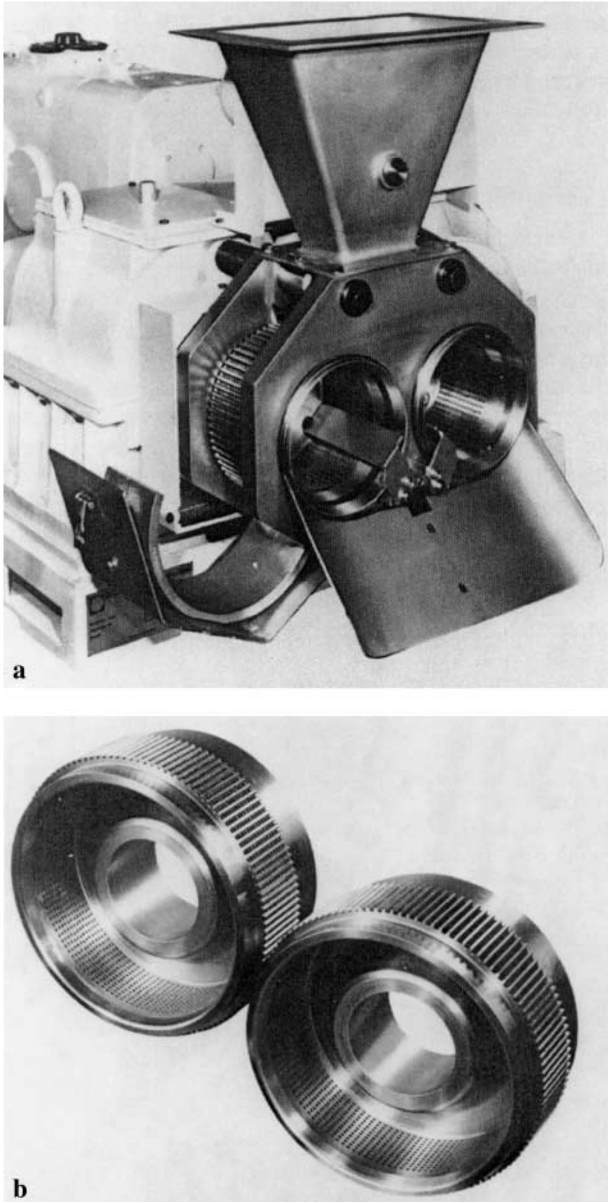

Fig. 13 (a) Rotary-gear extruder. (b) Gear-toothed cylinders.

Ram extruder

Industrial ram extruders are commonly used in the plastics and rubber industries for the preparation of warm strip feed for large cold-fed screw extruders and for forming strips or slugs for feeding injection molding and compression molding machines. They are used in the extrusion of specialized substances that require critical in-process control or that are not readily amenable to processing by screw extruders. Examples include the extrusion of waxlike substances such as coloring crayons, dental waxes, and rocket propellant, and in the extrusion of moist powders and claylike materials, such as blackboard chalks. Ram extrusion allows control of parameters, such as temperature, size, and weight of extrudate. An example of a high performance ram extruder, as manufactured by Borwell International Ltd., United Kingdom, is shown in Fig. 14. It consists of a chrome-plated barrel positioned in a thermostatically controlled storage tunnel. A range of dies can be fitted to the extruder head. Material is loaded into the barrel by manual or mechanical means and vacuum is applied to eliminate air from the system. The material is extruded by means of an hydraulically powered ram, with the hydraulic (oil) fluid being passed through a special valve system to sense changes in the plasticity of the material and compensate ram pressure to achieve an even extrusion through the hole. A multispeed cutter mounted on a fly wheel severs the extrudate at the die face. The volume of the extrudate is a function of the cut speed and the set ram speed, and can be controlled to a high degree of accuracy within $\pm 1\%$. A continuous operation is possible with the help of a twin barrel and ram arrangement in which material is fed to each barrel in turn by a screw system. Various sizes of extruder are available, from 4.5-L barrel capacity up to 80 L, offering production rates up to a maximum of 800 kg/h.

Choice of extruder

The selection of the extruder design is based on the principal requirements of the extrudate and the nature of further processing. For the production of uniform granules to be dried in a fluid-bed drier, a low-compaction system, such as that provided by the various types of screen extruders may be suitable. Cylinder or gear-type extruders may be more appropriate when aiming for a densified extrudate, such as that required for spheronization. Ram-extrusion systems, which allow precision control of extrudate density, size, and shape, are ideal for the extrusion and forming of pharmaceutical polymers of the type used for subdermal implants.

Consideration should be given to the availability of small-scale equipment, which is vital for development work prior to scale-up on pilot- or production-scale machines. Equipment choice is not necessarily based on maximum throughput rate, since the subsequent processing stages (e.g., cutting, spheronization, and drying) are batch processes and are therefore, a rate-limiting factor in production. Since extrusion is a continuous process, it allows adequate production rates for most purposes with any of the above mentioned extruder types.

The equipment must comply with the code of Good Manufacturing Practice (GMP) standards within the

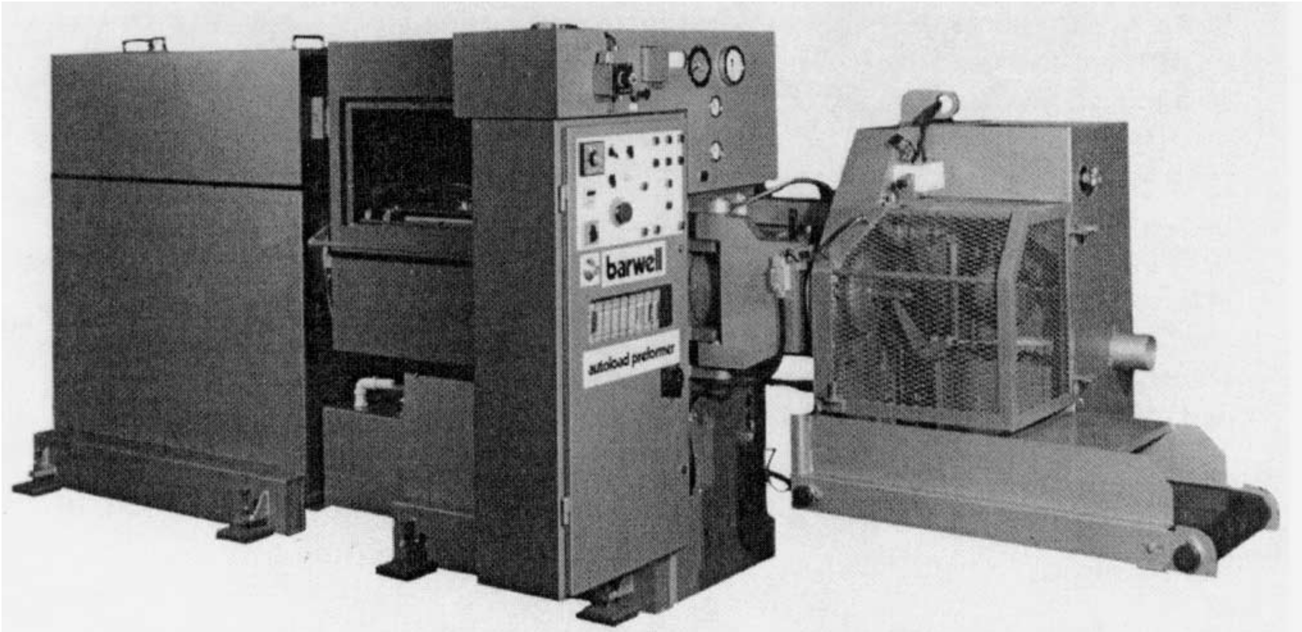

Fig. 14 The Barwell ram extruder.

pharmaceutical industry. Machines should be constructed of durable material with smooth surfaces to discourage adhesion of extraneous material and facilitate cleaning. Materials used for equipment construction must not affect the product. They should be corrosion resistant and able to withstand cleaning disinfectants. All surfaces and parts in contact with the product must therefore be of high-grade stainless steel and designed to exclude contamination of process material or product by lubricant during manufacture. Controls should be accessible by means of recessed contact buttons.

NOMENCLATURE

A_0	initial cross-sectional area
A	die cross-sectional area
C	compliance
D	die diameter
D_0	barrel diameter
$(dv/dr)_w$	rate of shear at the die wall
ESR	tensile stretch rate
EV	apparent elongational viscosity
K	power law viscosity constant
L	length of capillary (die)
n	degree of non-Newtonian flow (power law index)
n_b	Bagely entrance correction
P	pressure drop along die
P_e	die exit pressure
P_0	upstream pressure loss
P_{0_w}	pressure drop at zero velocity
P_T	total pressure drop
P_{t_v}	pressure drop at velocity v
Q	volumetric flow rate of extradate
R	radius of capillary (die)
RS	recoverable shear
T	tensile stress
U	plastic viscosity
V	extrudate velocity
x	thickness of Newton liquid layer
α	die entry yield stress velocity factor
β	die and velocity factor
γ	rate of shear
η	apparent viscosity
σ_y	yield value
σ_{y_0}	yield value at zero velocity
θ	half angle of convergence
τ	shear stress
τ_0	die wall shear stress at zero velocity
τ_w	die wall shear stress
τ_y	shear stress to be exceeded before flow commences

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